

Non-congruent phase transitions in cosmic matter and in the laboratory

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Non-congruence appears to be the most general form of phase transition in cosmic matter and in the laboratory. In terrestrial applications non-congruence means coexistence of phases with different chemical composition in systems consisting of two (or more) chemical elements. It is just the case for all phase transitions in high-temperature chemically reactive mixtures, which are typical for uranium-bearing compounds in many nuclear energy devices, both contemporary and perspective. As for cosmic matter, most of real and hypothetical phase transitions without nuclear reactions, i.e., those in the interiors of giant planets (solar and extrasolar), those in brown dwarfs and other sub-stellar objects, as well as in the outer crust of compact stars, are very plausible candidates for such type of phase transformations. Two exotic phase transitions, the gas-liquid phase transition in dense nuclear matter and the quark-hadron transition occurring in the interior of compact stars as well as in high-energy heavy-ion collisions are under discussion as the most extreme example of hypothetical non-congruence for phase transformations in High Energy Density Matter.

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1. Introduction

The term *non-congruent phase transition* (NCPT) denotes the situation of phase coexistence of two (or more) phases with different chemical compositions. This is a rather evident definition for the case of phase transitions (PT) in most of terrestrial applications (see below) and in astrophysical applications, where nuclear transformations, including β -decay, are negligible: PTs in planetary science and outer crust of compact stars etc. The nuclear composition in such situations is conserved and there is no problem with the selection of systems, which fulfill the condition of a NCPT. The situation is more complicated under extreme conditions like in the interiors

of compact stars and in remnants of supernova explosions, where nuclear transformations are close to equilibrium. The problem of the NCPT relevance is even more complicated in exotic situations with equilibrium hadron decay and quark deconfinement in interiors of strange (hybrid) stars and in the hypothetical quark-hadron (QH) phase transition in ultrarelativistic heavy-ion collisions at RHIC, LHC, FAIR and NICA. Hence the study of non-congruent phase transitions in typical terrestrial applications could be a useful base for understanding the relevance for such type of phase transitions in exotic situations like interiors of compact stars, supernova explosions, and in the hydrodynamic expansion of a fireball formed in heavy-ion collisions.

2. General features of non-congruent phase transitions in chemically reactive plasmas

Phase equilibrium in chemically reactive non-ideal plasmas of two or more chemical elements differs fundamentally from the case of ordinary phase equilibrium like, for example, the Van der Waals PT in substances with fixed chemical compositions (stoichiometry). Phase transitions in chemically reactive mixtures, including those in high-temperature uranium-bearing compounds, are typical for many nuclear energy devices both contemporary [23] and perspective [8, 19]. The basic feature of such two-phase systems is their non-congruency, i.e. their ability to vary stoichiometries of coexisting phases without violating the stoichiometry for the whole two-phase mixture. Non-congruency changes significantly the properties of all phase transitions in such systems, namely:

(A) - The significant impact of the *phase transformation dynamics*, i.e. of the strong dependence of the phase transition parameters on the rapidity of the transition. This dependence is of primary importance in experiments with fast surface evaporation of condensed samples under the powerful laser heating or electron-beam energy deposition. The strong competition between diffusion and thermal conductivity processes determines the parameters of such non-congruent evaporation;

(B) - The *phase transition thermodynamics* becomes more complicated. The essential changes include the scale of two-phase boundaries in extensive thermodynamic variables (say P - ρ etc) and even in topology of all two-phase boundaries in the space of intensive thermodynamic variables, as well as properties and even nature of the singular points (critical point first of all) and appearance of additional end-points in NCPT. One of the most remarkable consequences of the non-congruency is the change of the general form of the two-phase boundary in the pressure-temperature plane (see Fig. 1 below). A two-dimensional "banana-like" region appears in the NCPT instead of the well-known one-dimensional $P - T$ saturation curve

for ordinary (congruent) PTs. A next remarkable property for a NCPT is that isothermal and isobaric crossovers of the two-phase region are no longer coincide. The isothermal NCPT starts and finishes at different pressures, while the isobaric NCPT starts and finishes at different temperatures [14]. This property is crucial for the interpretation of the NCPT relevance in the physics of compact stars and high-energy heavy-ion collisions.

3. Conditions of joint phase, chemical and ionization equilibrium

3.1. Equilibrium between macroscopic phases with neutral species

3.1.1. Maxwell conditions

Phase equilibrium conditions for two macroscopic phases are well known for the case when coexisting phases consist of arbitrary mixtures of neutral species with equilibrium chemical reactions. In accordance with chemical thermodynamics laws these conditions include conditions of equilibrium heat and impulse exchange (equality for pressures and temperatures: $P' = P''$, $T' = T''$) and conditions of equilibrium matter exchange. The latter conditions have two variants for systems consisting of two or more chemical elements. The first one corresponds to *partial* equilibrium for exchange of matter with *fixed chemical composition*. This condition is equivalent to the well-known Maxwell "equal squares" construction for pressure-volume dependence in the case when both coexisting phases can be described by unique thermal equation of state (EOS) $P(V, T)$. For example, it is so for Van der Waals (gas-liquid) phase transition.

More general is the well-known "double tangent" construction for two free energies, $F'(V, T, x)$ and $F''(V, T, x)$ when both coexisting phases are described by different EOS-s. For example, it is so for crystal-fluid phase transition. In both the variants the final equilibrium condition corresponds to equality of Gibbs free energies of coexisting phases with fixed chemical composition:

$$T' = T'' , \quad P' = P'' , \quad G'(T, \rho', x) = G''(T, \rho'', x) \quad (1)$$

This form of phase equilibrium condition is often noted as "Maxwell condition" in astrophysical literature (for example [21]).

3.1.2. Gibbs conditions

The second variant corresponds to the *total* equilibrium in mean-phase matter exchange, i.e. equilibrium for exchange by each species with varying chemical composition of coexisting phases ($x' \neq x''$), but without violation of total chemical composition of whole two-phase system. This variant leads

to not one, but several separate equalities for partial quantities - chemical potentials μ_i ($i=1,2,\dots,k$ - all species) at $T' = T''$ and $P' = P''$

$$\mu'_i(T, \rho', x') = \mu''_i(T, \rho'', x'') , \quad \alpha x' + (1 - \alpha)x'' = x \quad (2)$$

In terrestrial applications this form of phase equilibrium conditions corresponds exactly to the definition of non-congruent phase transition. For the case of equilibrium chemical reactions in each phase total number of equalities for chemical potentials is decreased to reduced number of equalities for chemical potentials of basic (independent) species. For example, it is two basic units, oxygen and uranium chemical potentials μ_{O} and μ_{U} , in the case of equilibrium uranium-oxygen mixture (see below). In astrophysical application the form (2) is well known under the name "Gibbs conditions". The problem is that this form is applied there to charged species, but not only to neutral ones (see below).

3.2. Phase equilibrium of macroscopic phases in presence of charged species (Gibbs - Guggenheim conditions)

Phase equilibrium conditions for macroscopic phases with charged species are more complicated. There are two basic points. The first one is that electroneutrality conditions are added for both phases in (1, 2). Maxwell conditions (1) are still valid for Gibbs free energies, G' and G'' , of *electroneutral* phases with chemical and ionization equilibrium inside. As for the Gibbs conditions (2), the point is that besides electroneutrality restrictions two additional quantities appear in description of coexisting phases and, correspondingly, in equilibrium conditions as additional independent variables. It is average electrostatic potentials, $\varphi'(r)$ and $\varphi''(r)$ [5] (see for example [11]). As a result, a remarkable feature of any Coulomb system is the existence of two versions of chemical potential, μ_i and $\tilde{\mu}_i$. The (ordinary) chemical potential, $\mu_i(n_k, T)$, is presumed to be a local parameter depending on local density, temperature and composition. The new (generalized) *electro-chemical potential* $\tilde{\mu}_i$ is not local parameter. It strongly depends on non-local sources of influence, such as total charge disbalance including surface dipole, other surface properties etc. In *uniform* Coulomb system $\tilde{\mu}_i$ is equal to the sum of $\mu_i(n_k, T)$ and average (bulk) electrostatic potential, φ , which is *presumed* to be *uniform* also.

$$\tilde{\mu}_i = \mu_i(\{n(\mathbf{r})\}, T(\mathbf{r})) + Z_i e \varphi(\mathbf{r}) \quad (3)$$

For each charged specie in Coulomb system the values of its chemical potentials in coexisting phases, μ'_i and μ''_i , must not be equal under conditions of phase equilibrium. It is namely the electro-chemical potential, to have the same values in coexisting phases at phase equilibrium:

$$\mu'_i(\{n'\}, T) \neq \mu''_i(\{n''\}, T) , \quad \tilde{\mu}'_i(\{n'\}, T) = \tilde{\mu}''_i(\{n''\}, T) \quad (4)$$

This form of phase equilibrium conditions (3, 4) we will refer below as Gibbs-Guggenheim conditions. Equalities (3, 4) being combined with the electroneutrality conditions leads to remarkable feature of any equilibrium Coulomb system, namely: every phase boundary in such system is accompanied, as a rule, by a finite gap in the average electrostatic potential through the phase interface [10, 11].

$$\Delta\varphi \equiv \varphi''(r \rightarrow +\infty) - \varphi'(r \rightarrow -\infty) = (\mu''_e - \mu'_e)(e)^{-1} = (\mu'_i - \mu''_i)(Ze)^{-1} \quad (5)$$

In contrast to the work function this inter-phase (Galvani) potential drop $\Delta\varphi$ represents a thermodynamic quantity, which does depend on temperature and chemical composition only and does not depend on surface properties. This gap tends to zero at the critical point of gas-liquid phase transition. The zero-temperature limit of this drop (along the coexistence curve) can be considered as an individual thermo-electrophysical coefficient of any material. The value of discussed potential drop could be directly calculated by numerical modeling of phase transitions in the Coulomb system when both the coexisting phases being explicitly simulated [11].

It should be stressed that any phase transition in plasmas of one chemical element, for example evaporation in metals, must be forced-congruent in spite the fact that one (or both) coexisting phases being composed of two basic units: ions and electrons (all other species being their equilibrium bound complexes). It is electroneutrality conditions in both (macroscopic) phases that make this coexistence thermodynamically one-dimensional. On the contrary, this system became two-dimensional (and all phase transitions became non-congruent) just at the moment when we relax electroneutrality conditions in both phases and allow equilibrium mean-phase exchange by charged species also. This is just the case in so-called "mixed phase" scenarios (see below).

3.3. Mesoscopic scenarios for phase equilibrium ("mixed phase" concept)

There exists very popular and widely accepted scenario for phase transition, which differs essentially from the both described above ones. The basic idea, which was claimed in [20] and developed in [3] and many other papers (for example [4]), is that in many astrophysical situations a highly dispersive, uniform and heterogeneous mixture of charged micro-fragments of one phase in oppositely charged "see" of another one (charged emulsion) may be more thermodynamically favorable (i.e. not metastable, as it is

in most of terrestrial applications (mist, foam etc.) but stable) than standard (Maxwell) form of forced-congruent coexistence of two electroneutral macroscopic phases.

The simplest form of mixed-phase equilibrium conditions is equivalent exactly to equations (2) as if all charged species are equilibrated in mean-phase exchange as well as neutrals species. In this simplest approximation all thermodynamic loss in such charged emulsion due to Coulomb energy of charge separation and positive contributions of surface tension are neglected.

More sophisticated form of discussed mesoscopic scenario for phase coexistence ("structured mixed phase", see for example [16]) takes into account mentioned above thermodynamic loss due to surface tension and charge separation. It leads to existence of optimal size, form and charge for micro-fragments of both mixed phases in mentioned above charged emulsion ("pasta plasma"). The question of degree of equivalence for 'structured mixed phase' and non-congruent PT is open. See discussion below.

4. Non-congruent evaporation in the uranium-oxygen system

Development of wide-range equation of state(EOS) for uranium and uranium-bearing compounds with taking into account all phase transformations in such systems, was the subject of multi-annual theoretical study [19, 23]. The physics of phase transitions in uranium dioxide ($\text{UO}_{2\pm x}$) is of primary importance for prediction of behavior of nuclear reactors during hypothetical severe accidents [23]. In set of the works [9, 14, 15] an adequate theoretical model of non-congruent evaporation in U-O system was developed. The basic point of the model is the description of both coexisting fluid phases (liquid and vapor) in a uniform manner, as equilibrium multi-component strongly interacting (non-ideal) mixtures of atoms, molecules, molecular and atomic ions, and electrons as well ("chemical picture", see e.g. [19]). Chemical reactions and ionization as well as the parameters of phase equilibrium have been calculated self-consistently by taking into account all significant non-ideality corrections (strong Coulomb interaction, intensive short-range repulsion and attraction) within modified version of thermodynamic theory. Details of the adopted approximations are described elsewhere [14, 23]. The fluid model (common for liquid and vapor phases) has been applied for self-consistent calculations of non-congruent phase coexistence within the wide range of temperature and pressure ($T \leq 20$ kK, $P \leq 2$ GPa) including the vicinity of the true critical point of non-congruent PT. The basic point of these calculations is that the Gibbs conditions (2) have been used for all neutral species in both phases, while the Gibbs - Guggenheim conditions (3,4) have been used for all charged species, the conditions, which have been actually violated in all previous studies of evaporation in

U-O system (for example [1]). The pressure-temperature phase diagram for non-congruent evaporation is shown in Fig. 1 as the most important result for present discussion.

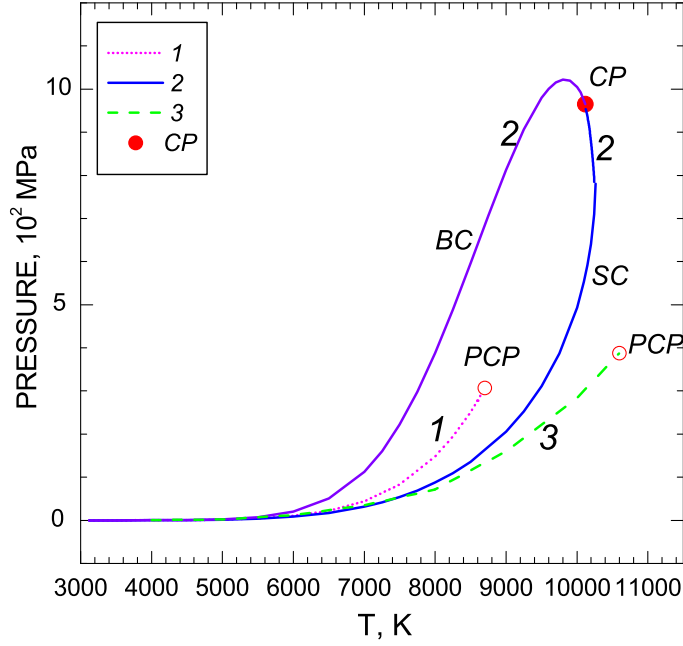


Fig. 1. Pressure-temperature diagram for non-congruent evaporation in chemically reacting U-O plasma ($O/U = 2.0$) [14]. 1,2– calculations via EOS [14]: 1– forced-congruent coexistence (Maxwell conditions (1)), *PCP* – pseudo-critical point. 2–2– boundaries of the two-phase region via total (non-congruent) equilibrium Eqs. (2)-(5): *BC*– boiling curve (bubble point), *SC*– saturation curve (dew point), *CP*– true critical point; 3– gas-liquid coexistence curve calculated via previous EOS of UO_2 [1].

5. General nature of non-congruent phase coexistence in compounds and chemical mixtures

Mentioned above long-time study of non-congruent phase equilibrium in U-O system [14, 15] indicates that this type of phase transformation is not as infrequent at high temperatures as it was seen before. The main conclusion drawn from above results could be formulated in following statement:

Any phase transition in equilibrium system containing two or more chemical elements must be non-congruent in general. Congruent phase transitions in such systems arise as exception only.

This statement seems to be in evident contradiction with our everyday experience because one knows very many examples of PT-s in compounds of two (or more) chemical elements, for example, in simple water and other substances (H_2O , CO_2 , NH_3 etc), where parameters of PT are studied exhaustively and nobody ever heard about non-congruence and banana-like P - T diagrams. Nevertheless, there is no contradiction. Gas-liquid PT in all these compounds are exceptions. All these PT-s are indeed congruent in *room conditions* because all of them conserve mono-molecular composition through the evaporation ($\text{H}_2\text{O} \rightleftharpoons \text{H}_2\text{O}$) and there is no any degree of freedom for two-phase system to change stoichiometry in liquid and/or vapor phases. But situation is absolutely different for PT-s in these compounds in planetary conditions ($T \sim 10$ - 20 kK, $P \sim 1$ - 10 Mbars). Expected nomenclature of PT-s in such conditions is very abundant (see for example [18]) while all discussed compounds are no more mono-molecular. Our present knowledge of parameters for these PT-s is very poor [6]. But qualitatively the main statement of present work is that any phase transition in these compounds in planetary conditions must be non-congruent, i.e. all P - T (or μ - T) boundaries for phase transitions must be two-dimensional regions instead of ordinary one-dimensional curves [15].

Generally, the expected examples of non-congruent phase transitions in terrestrial applications are inter alia:

- Uranium- and plutonium-bearing compounds: ($\text{PuO}_{2\pm x}$, UC, UN etc),
- Evaporation in other oxides (for example, in SiO_2)
- Evaporation in hydrides of metals (for example, in LiH)
- Evaporation in ionic liquids and molten salts: (for example, in NaCl)
- Evaporation in metallic alloys
- Phase transitions in "dusty" and colloid plasmas: (Coulomb system of macro- and micro-ions with charge $q_M = +Z, q_m = \pm 1$)

6. Non-Congruence in cosmic matter

6.1. Ordinary situations

There exist many candidates for such type of phase transitions in cosmic matter without nuclear transformations:

- Hypothetical plasma- and dissociation-driven phase transitions in mixture $\text{H}_2 + \text{He}$ (+ $\text{H}_2\text{O} + \text{NH}_3 + \text{CH}_4$) in interiors of giant planets (Jupiter, Saturn, Neptune etc), in brown dwarfs and in extra-solar planets [15],
- Phase transitions in isentropically released products of strong shock compression of lunar ground ($\text{SiO}_2 + \text{FeO} + \text{Al}_2\text{O}_3 + \text{CaO} + \dots$) under huge impact after natural (meteorite) or artificial (LCROSS mission) bombarding
- Crystallization and ionic demixing in interiors of white dwarfs,
- Crystallization and ionic demixing in outer envelopes of compact stars (for example [7]).

6.2. Non-congruence in exotic situations

Relevance of non-congruent scenario for phase transition in exotic situations is not transparent. There exist many phase transitions, which could be considered as candidates for such transformations. Two groups of them will be commented here as the first ones:

(I) Gas-liquid (Van der Waals-like) phase transition in dense nuclear matter of equilibrium mixture of p, n, e and nuclei $\{N(A, Z)\}$. Here $\{N(A, Z)\}$ is equilibrium ensemble of all possible bound complexes from Z protons and $(A - Z)$ neutrons (see [22] and reference therein). Several variants may be considered: with and without electrons, electroneutrality and Coulomb interaction, and with and without β -equilibrium).

(II) Hypothetical phase transition(s) in the vicinity of quark deconfinement boundary at high temperature and with very complicated nomenclature of hypothetical phase transformation at relatively low temperature (for example [4]).

6.2.1. Gas-liquid phase transition

(I.a) Gas-liquid (GL) phase transition (PT) in equilibrium mixture $\{p, n, N(A, Z)\}$ with no electrons, no electroneutrality, no Coulomb repulsion (for example [22, 2]). The system is equivalent to chemically reacting mixture of two chemical elements. The symmetry parameter Y - is independent variable. It is equivalent to stoichiometry (chemical composition) in ordinary chemical mixtures. Hence, this GLPT is non-congruent in non-symmetric case ($Y \neq 0.5$) and congruent (i.e. azeotropic) in symmetric case ($Y = 0.5$).

(I.b) Ordinary GLPT (with macroscopic coexisting phases) in the same mixture $\{p, n, e, N(A, Z)\}$ with electrons, Coulomb interaction and electroneutrality and with β -equilibrium. The system is equivalent to *one-*

dimensional (thermodynamically) system like partially ionized and dissociated hydrogen $\{p, e, H, H_2, H^-, H_2^+ \dots \text{etc.}\}$ (Gibbs-Guggenheim conditions (3-5)). Hence, this GLPT is forced-congruent (VdW-like).

(I.c) GLPT in the same mixture as in (I.b) $\{p, n, e, N(A, Z)\}$ in frames of simplest mesoscopic scenarios (simple "mixed phase"). No local electroneutrality, only global one. (Gibbs conditions (2)). This system is again equivalent to two-component (two-element) chemically reacting terrestrial mixture. Hence, this GLPT is non-congruent in general. $P - T$ and $\mu - T$ phase boundary must be two-dimensional banana-like region instead of ordinary (VdW-like) saturation curve.

(I.d) GLPT in the same mixture as in (I.c) in frames of advanced mesoscopic scenarios ("structured mixed phase" - "pasta plasmas"). The most complicated situation. This system is not equivalent to any terrestrial analog. Problem of congruence for such GLPT should be analyzed separately.

6.2.2. Quark-hadron phase transition

(II.a) Quark-hadron (QH) phase equilibrium (PT) between macroscopic quark-gluon and hadron phases is one-dimensional (thermodynamically) system. Phase transitions must obey to the Gibbs-Guggenheim conditions (3-5). Hence this variant of QHPT is equivalent to congruent PT, i.e. $P - T$ and $\mu - T$ phase boundaries must be one-dimensional curves rather than two-dimensional stripes. It should be stressed that this variant of QHPT is not equivalent to VdW-like PT (like case I.b) by two reasons. First, this variant of QHPT is much closer to entropic type of PT (i.e. decreasing $P - T$ coexisting curve, small density gap etc.) than to enthalpic one like VdW-PT (i.e. increasing $P - T$ coexisting curve, large density gap etc.) [12] [13]. Second, presently considering versions of QHPT are described by separate analytic EOS-s for quark and hadron phases. Hence, there is no reason to expect appearance of critical point in such descriptions like it is in the case of crystal-fluid phase transition in terrestrial physics [17].

(II.b) QHPT in the same combination as in (II.a) in frames of simplest *mesoscopic* scenarios (simple "mixed phase"). No local electroneutrality, only global one. Gibbs conditions (2) are valid for all species, charged and neutral. Quark-hadron phase transition via "mixed-phase" scenario has the main features of non-congruent phase transitions: Isothermal transitions through the two-phase region starts and finishes at *different pressures* (and at *different partial chemical potentials*). This system is equivalent to two-dimensional (thermodynamically) system. Hence, this version of QHPT is non-congruent in general. $P - T$ and $\mu - T$ phase boundaries are two-dimensional stripes rather than one-dimensional curves.

(II.c) QHPT in the same combination as in (II.a) in frames of advanced mesoscopic scenario: "structured mixed phase" - "pasta plasma" (for exam-

ple [16]). The most complicated situation. This system is not equivalent to any terrestrial analog. Problem of congruence or non-congruence for such QHPT should be analyzed separately.

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